Transport properties of a two Co atoms system: a theoretical approach.

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(Dated: September 18, 2012)

A system of two interacting Co atoms has been studied in a recent Scanning Tunneling Microscope experiment by J. Bork *et al.*, Nature Physics **7**, 901 (2011). This kind of system has been intensively analyzed since a quantum critical point was theoretically predicted for an antiferromagnetic interacting dimer. We propose a two path structure microscopic model capable of explaining, in all the parameter space, the physics of this experiment. It consists of two atoms interacting directly and indirectly through a hopping between two electronic reservoirs that bypasses the atoms. The many-body system is treated in the finite-U Slave Boson Mean Field Approximation. Other results obtained using the Logarithmic Discretization Embedded Cluster Approximation are also discussed. The system is characterized as being in the Kondo regime for all the parameter space experimentally studied, although the effective exchange interaction between the Co atoms is antiferromagnetic.

PACS numbers: 73.23.Hk, 72.15.Qm, 73.63.Kv

Introduction.—The two-impurity Kondo model (TIKM) and the closely related two-impurity Anderson model (TIAM) are two of the simplest strongly correlated models containing the essential physics necessary to study a system that undergoes a quantum phase transition (QPT) between a phase characterized by two independent Kondo-screened magnetic impurities and another where the two impurities are locked into a dimer, forming a spin singlet state [1]. The parameter that drives this phase transition is the ratio I/T_K^0, where I is the inter-impurity exchange interaction and T_K^0 is the Kondo temperature of the individual impurities (assumed to be identical). Experiments performed in two-impurity systems [2, 3] have been able to carry the system from the Kondo screened phase to the antiferromagnetic (AF) regime, but without a precise control of the exchange interaction between the two impurities that is essential to observe the fluctuations associated to the quantum critical point (QCP) corresponding to the QPT mentioned above. Furthermore, it is well known that both the TIKM [4–8] and the Two Channel Kondo Model (TCKM) [8, 9] are easily driven out of their respective QCPs by inter-lead hopping perturbations and for this reason it has been very difficult to experimentally observe signals of quantum criticality. To the best of our knowledge, the only known exception is the TCKM studied by Potok *et al.* [10]. Moreover, recent theoretical works have suggested that these classes of QPTs are not observable under current experimental conditions [11, 12]. Nevertheless, alternative setups have been theoretically proposed [13] to try and observe them.

Recently, a remarkable experiment has been performed where a Co atom, positioned at the tip of a Scanning Tunneling Microscope (STM), is continuously approached to another Co atom adsorbed on an Au(111) surface [14]. The position of the STM tip was varied with sub-picometer (pm) accuracy and hence the ratio I/T_K^0 could be modified almost continuously. However, as the tip approaches the surface, the *direct* charge transfer between them increases, thus driving the two-impurity system away from the QCP [15, 16]. In Ref. [14], the system stayed in a regime away from the neighborhood of the QCP, as a peak in the differential conductance (dC), expected to appear at the QPT [16, 17], was never observed. This fact was interpreted as resulting from the direct hopping between the electrodes. Some features observed in the experiments, namely the splitting in dC at small tip/surface separations, could be explained by a microscopic model that takes into account only the hopping between the electronic reservoirs [14] and which was solved using the Numerical Renormalization Group (NRG) method [18]. This splitting was interpreted as originating from the effective exchange interaction between the Co atoms [19–21]. However, other experimental features, such as Fano anti-resonances that first become narrower and then transform into a peak, before splitting when z is reduced, have not been explained using a *microscopic* model [22]. Besides, it is important to physically characterize the system as the distance between impurities is decreased.

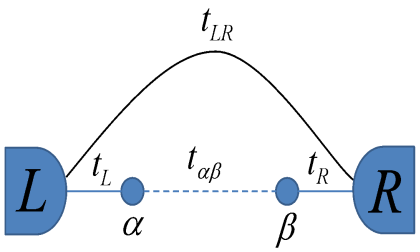

FIG. 1: (Color online) Schematics of the model studied in this work. α and β represent the two Co atoms.

The authors show in this work that the double-Co experiment described above can be interpreted and understood, for all inter-impurity distances studied, by a model that also incorporates, as an essential ingredient, *a direct hopping between*

the Co atoms. In particular, the results show to what extent the interplay between the direct inter-impurity hopping and the indirect one influences the transport properties of the system.

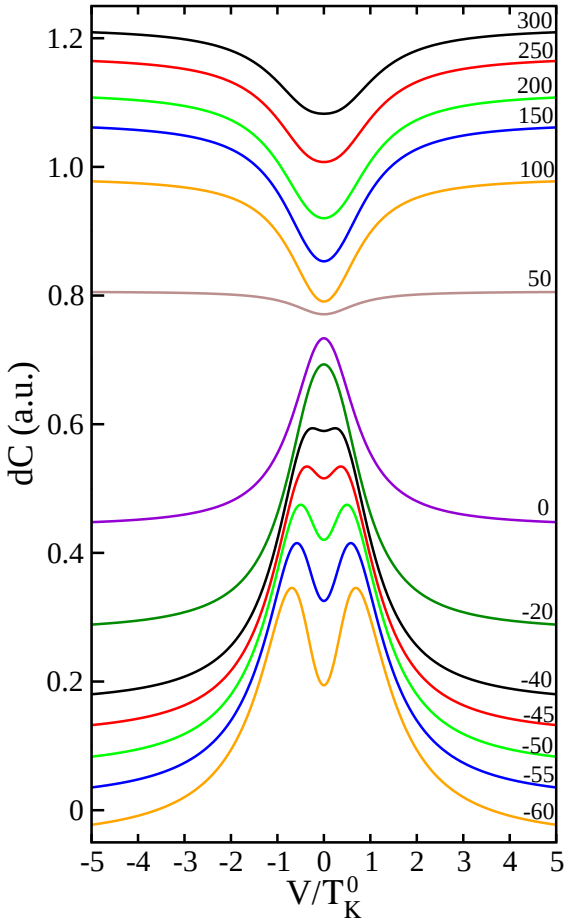

FIG. 2: (Color online) Differential conductance as a function of V/T_K^0 for different values of z (separation between the impurities, measured in pm, indicated in the right side). Each curve, calculated using SBMFA, has been shifted vertically for clarity.

Model.—The Hamiltonian is written as a sum of three terms, namely

$$H = H_{\rm imp} + H_{\rm hyb} + H_{\rm leads}, \tag{1}$$

where

$$H_{\rm imp} = \sum_{i=\alpha,\beta;\sigma} \left(\epsilon_i n_{i\sigma} + \frac{U}{2} n_{i\sigma} n_{i\bar{\sigma}} \right) \tag{2}$$

describes the isolated impurities, ϵ_i (where $i = \alpha,\ \beta$) being the energy of each localized impurity, U the on-site Coulomb interaction, and $\sigma = \pm$ is the spin orientation. The contribution

$$H_{\rm hyb} = \sum_{\sigma} t_L c^{\dagger}_{L,1\sigma} c_{\alpha\sigma} + t_R c^{\dagger}_{R,1\sigma} c_{\beta\sigma} + t_{\alpha\beta} c^{\dagger}_{\alpha\sigma} c_{\beta\sigma} + t_{LR} c^{\dagger}_{L,1\sigma} c_{R,1\sigma} + \text{H.c.} \tag{3}$$

describes the hybridization of each impurity with the first site of its adjacent metallic lead (modeled as semi-infinite non-interacting chains, see below), the hybridization between both impurities and the direct tunneling between the left (L) and right (R) electron reservoirs (in that order). Finally,

$$H_{\rm leads} = t \sum_{j=L,R} \sum_{i=1;\sigma}^{\infty} \left(c^{\dagger}_{j,i\sigma} c_{j,i+1\sigma} + \text{H.c.} \right), \tag{4}$$

describes the L, R leads, represented by two semi-infinite chains of non-interacting sites with hopping t between adjacent sites. The model is depicted in Fig. 1. Most of the transport properties were calculated within the finite-U Slave Bosons Mean Field Approximation (SBMFA) [23], although, for the sake of comparison, some results were obtained using the Logarithmic Discretization Embedded Cluster Approximation (LDECA) [24]. Finally, for the sake of simplicity, we adopt a symmetric model (i.e., $t_L = t_R = t'$) [25].

We define T_K^0 as the Kondo temperature for each Co atom in the two independent single-impurity Anderson models that result from taking $t_{LR} = t_{\alpha\beta} = 0$, where T_K^0 is obtained through the half-width of the impurity local density of states (LDOS) at the Fermi energy. Taking, in units of t, $U = 0.8$, $t' = 0.25$, and $\epsilon_i = -U/2$, we obtain $T_K^0 = 0.0073$. Considering $t = 1.3$ eV for Au, one obtains $T_K^0 \approx 9.5$ meV $\equiv$ 110 K, roughly the same Kondo temperature measured in the experiments with Co atoms on Au [14]. T_K^0 will be a reference for comparison with experiments. The values of U and t' were chosen, within the convergence parameter space of the SBMFA method, so that $U/\Gamma \sim 14$, where $\Gamma = \pi t'^2 \rho(E_F)$, being $\rho(E_F)$ the leads' DOS at the Fermi energy. This U/Γ value assures that the single impurity system is deep inside the Kondo regime at zero temperature.

Our model includes the hopping t_{LR} between the electron reservoirs, providing a channel through which the dots couple indirectly and also a weaker direct hopping $t_{\alpha\beta}$ between the Co atoms that, as mentioned above, results to be essential to reproduce the experimental results. The assumption that $t_{\alpha\beta} \ll t_{LR}$ is in accordance with the hypothesis made in Ref. [14], namely, that for a vertical approach between the STM tip and surface, the interaction between the d-orbitals of the Co atoms is very weak. Assuming the hoppings to follow $t_{LR} = Ae^{\gamma z}$ and $t_{\alpha\beta} = Be^{\delta z}$, where z is a parameter representing the inter-impurity distance, then a decrease in z results in an exponential increase in $t_{\alpha\beta}$ and t_{LR} (for $\gamma,\ \delta < 0$). After an extensive survey, by varying the parameters so as to take into account the constraint $t_{\alpha\beta} << t_{LR} < t$, as discussed above, and the obvious conditions $t_{\alpha\beta} \approx 0$ and $t_{LR} << 1$ for the largest z-value used, we found that the parameters that best allow the reproduction of the experimental results are $A = 0.4066$, $B = 0.0305$, $\gamma = -0.002534$, and $\delta = -0.00973$ (compare our Fig. 2 with Fig. 4(b) in Ref. [14]). For these parameter values, t_{LR} varies from 0.19 to 0.47 and $t_{\alpha\beta}$ from 0.0015 to 0.055, in the interval $-60 \leq z \leq 300$ (same as in the experiment, in pm units). This parametrization allows us to reproduce qualita-

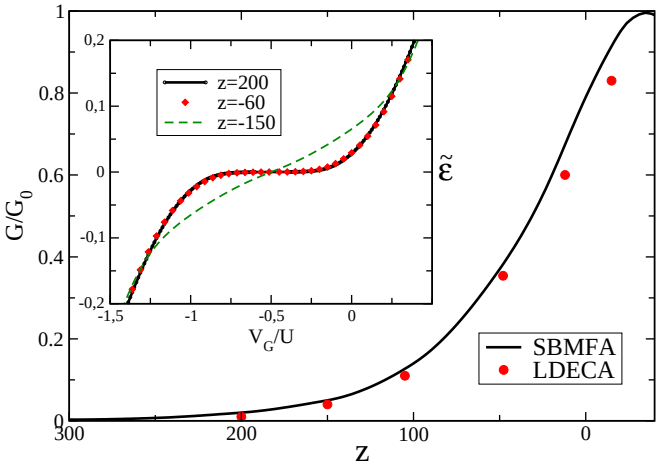

FIG. 3: (Color online) Main panel: Conductance G/G_0 as a function of z. When compared to Fig. 4(a) in Ref. [14], we see the same overall behavior. Inset: renormalized energy level $\tilde{\epsilon}_\alpha = \tilde{\epsilon}_\beta = \tilde{\epsilon}$ as a function of gate voltage V_g. The plateau at the Fermi energy for both $z = 200$ (continuous (black) line) and $z = -60$ [(red) diamonds line], indicates that the system stays in the Kondo regime. For larger hopping values, $t_{\alpha\beta} = 0.06$ and $t_{LR} = 0.13$, the plateau starts to be suppressed, as shown by the (green) dashed curve for $z = -150$, reflecting that the system enters a crossover regime (see text)

tively and semiquantitatively the experimental results for dC for the whole sequence of z values.

In the SBMFA, dC is calculated, using the Keldysh formalism [26]. For simplicity, without an applied potential, we assume electron-hole symmetry. The effect of an external bias V is taken into account supposing that the system is connected to a left and right reservoirs, which Fermi levels are $V/2$ and $-V/2$ respectively. In this case, the differential conductance can be written as, $dC = 4\pi^2 t'^4 \mathrm{Im}\{G_L(V/2)\}\mathrm{Im}\{G_R(V/2)\}|G^V_{LR}(V/2)|^2$, where $G_L = G_R$ are the reservoirs' non-interacting Green's functions and $G^V_{LR}(\omega)$ is the many-body propagator from left to right reservoirs, under the presence of V. To calculate this Green function, within the SBMFA formalism, the renormalized parameters of the Hamiltonian have to be self-consistently calculated incorporating the external bias. However, it is known [27] that for a two-impurity system the self-consistent results obtained for the equilibrium situation ($V = 0$) are very similar to the non-equilibrium results (finite V), as long as V is smaller than a few times T^0_K and $T^0_K \lesssim t_{\alpha\beta} \lesssim t^2_{\alpha\beta}/U$. Under these conditions, the propagator $G^V_{LR}(\omega)$ is almost independent of the external bias V. Therefore, we will assume its complete independence from V and calculate dC as if the system were in equilibrium [28].

Results.—In Fig. 2, we present the SBMFA results for dC as a function of V/T^0_K. The dC curves present three fundamental features that should be emphasized: *(i)* for negative values of z, there is a double-peak structure, displaying a splitting that decreases with increasing z; *(ii)* eventually, still for negative values of z, the splitting is totally suppressed, becoming a single peak; *(iii)* for higher z values, a Fano anti-resonance develops centered at $V = 0$, with increasing width as z increases (see also Fig. 4). The behavior just described is qualitatively and semiquantitatively similar to that observed in the experiments by Bork *et al.* (Fig. 4(b) of Ref. [14]). However, the range of z values for which our results show a single-peak is slightly larger than in the experiments, where an anti-resonance line shape persists down to $z \approx -30$, while our results show a single-peak feature already at $z = 0$.

In Fig. 3 we present SBMFA (dark line) and LDECA (solid (red) dots) results for the conductance G/G_0 as a function of z (where $G_0 = 2e^2/h$ is the quantum of conductance), which can be compared to Fig. 4(a) in Ref. [14]. Since LDECA is exact at the Fermi energy [24], the excellent agreement between SBMFA and LDECA gives support to the SBMFA results shown in this paper. The main difference with the experimental results is that our G/G_0 values increase more smoothly as z decreases. This can be associated to the fact that, as expected, the experimental results are very dependent upon the distance between the atoms. As stated by Bork *et al.*, 'mechanical relaxation' effects should be at play when a transition from 'tunneling' to 'point contact' occurs, as the tip gets closer to the surface. Hence, at this transition the real distance z and the associated hopping parameters $t_{\alpha\beta}$ and t_{LR} are difficult to determine. Besides, other matrix elements such as t_L or t_R, assumed to be constant, may vary at this transition.

The LDOS of the impurities calculated with SBMFA (not shown) shows the widening of the Kondo resonance as z decreases. This is in accordance with previous studies in similar systems [30]. In addition, the splitting in dC is directly correlated to an equivalent splitting in the impurities' LDOS. Note that a split-peak in dC was also obtained by using NRG [14], with a model where only the hopping between the electronic reservoirs was included. However, the overall agreement between our theoretical results and the experiments requires the inclusion, as a crucial parameter, of the *direct* inter-impurity interaction $t_{\alpha\beta}$. In addition, using SBMFA, we have calculated the phase difference (not shown) between the two channels through which the current can flow, namely the channel passing directly through the impurities (via $t_{\alpha\beta}$) and the channel connecting the electronic reservoirs directly (via t_{LR}). We obtained that the phase difference is zero for all values of z, indicating that these two channels interfere constructively.

In the SBMFA, when the system is in the Kondo regime, there is a plateau at the Fermi energy ($E_\mathrm{F} = 0$) in the renormalized energy level of each impurity ($\tilde{\epsilon}_\alpha = \tilde{\epsilon}_\beta = \tilde{\epsilon}$), as a function of gate voltage V_g [23]. As can be seen in the inset of Fig. 3, for the region of interest, the plateau is perfectly defined (compare (black) solid curve for $z = 200$ with the almost identical (red) diamonds curve for $z = -60$), which leads to the conclusion that the system remains in the Kondo regime for this range of z. It is important to emphasize that, although the system remains in the Kondo regime, the splitting in dC is relatively large for the lowest values of z, as shown in Fig. 4, discussed below. For larger values of $t_{\alpha\beta}$ and t_{LR}, the plateau is partially suppressed as shown in the (green)

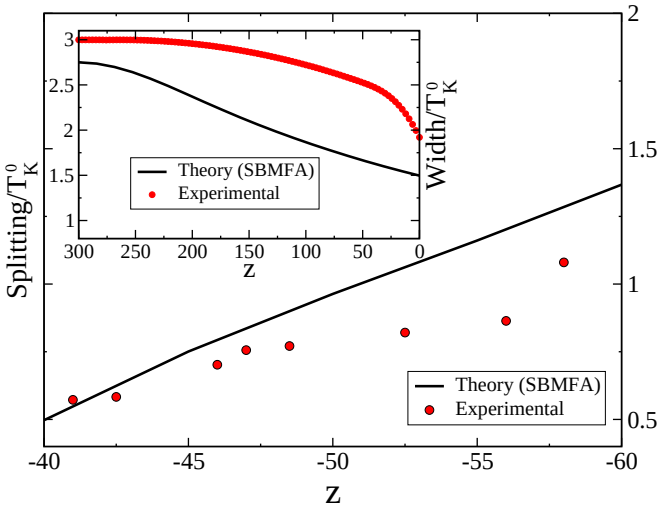

FIG. 4: (Color Online) Splitting as a function of z[(black) solid curve]. Inset: Width of the anti-resonance as a function of distance in units of T_K^0. The SBMFA results are similar to those obtained in Figs. 5(b) and 5(a) in Ref. [14] [here reproduced schematically as solid (red) dots].

dashed curve for $z = -150$. Thus, for large hopping values the system enters a crossover region, which is compatible with measurements presented in Fig. S.6 of Ref. [14], transitioning from the Kondo to an inter-atomic singlet state. This indicates that the direct and indirect couplings between the impurities result in an effective antiferromagnetic spin-spin correlation between them, suppressing the Kondo state in each impurity.

The main panel in Fig. 4 shows the dC-splitting (distance between peaks) as a function of z (in the interval $-60 \leq z \leq -40$). The (black) solid curve represents the SBMFA results and the (red) solid dots are the experimental results extracted from Ref. [14], both in units of their corresponding single-impurity Kondo temperature T_K^0, which is approximately $75\ K \approx 6.5\ meV$ in the experiments. The (black) solid curve shows that the splitting has an almost linear dependence with z. In the inset of Fig. 4, we show the SBMFA [(black) solid curve] and the experimental [(red) solid dots curve] results for the width of the dC Fano anti-resonance as a function of z, in the interval $300 \geq z \geq 0$, scaled by T_K^0. We fit the SBMFA dC curve with a Fano anti-resonance to extract its width. The sharper decrease in the dip width for the experimental results at $z \approx 25$ pm can be ascribed to the relaxation process that takes place, changing the distance between the Co atoms, as mentioned above. We stress the fact that as the magnitude of the splitting can change when the experiment is repeated, a qualitative and semiquantitative description of the experimental results should be satisfactory.

Single impurity case.—In the case of a single Co impurity, the experimental dC shows only one dip, which neither diminishes its width nor transforms into a peak as the distance between tip and surface is reduced [14, 31]. In order to study the differences between the single- and double-impurity cases, we have carried out a z dependence study for the single-impurity model. The model is depicted in the inset of Fig. 5. The hopping between the impurity and one of the reservoirs (t_R) and the hopping between the two reservoirs (t_{LR}) are varied in the same way as in the double-atom case. The SBMFA results (main panel of Fig. 5) show an asymmetric anti-resonance, as previously obtained [32]. We checked that the dip's width does not decrease by changing z. Besides, the dip reaches its minimum at negative values of bias voltage, as noted in Ref. [32] for an $S = 1/2$ impurity, which is our case. We note that Co atoms have $S = 3/2$, but, as mentioned in Ref. [32], there are only quantitative differences between the $S = 1/2$ and $S = 3/2$ cases. The value of G/G_0 at the Fermi level is much smaller than in the two-impurity case. Therefore, these results show that the reduction of the anti-resonance width, the appearance of a peak and its splitting, as was observed in the experiments of Ref. [14], are a consequence of the presence of a second impurity, interacting with the first.

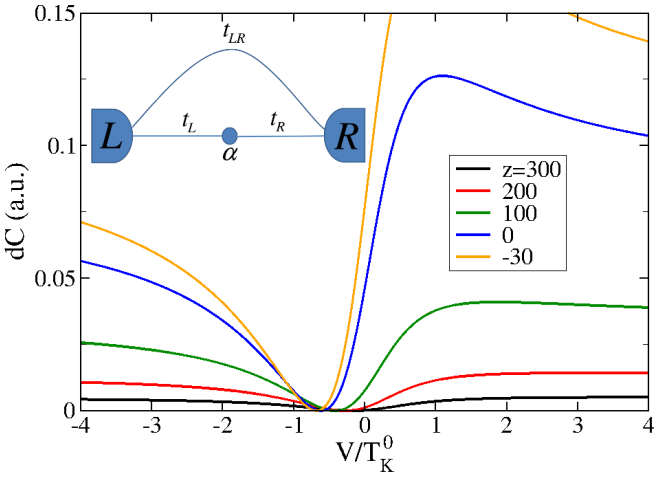

FIG. 5: (Color Online) Differential conductance for a single impurity as a function of V/T_K^0. The behavior is markedly different from the case of two atoms, as in the experiments.

Conclusions.—Summarizing, the model proposed in this work to study the transport properties through two Co atoms in series correctly describes the behavior observed in STM experiments for all the parameter range. In that respect, the inclusion of a direct hopping between the atoms, *besides* the one between the electron reservoirs, proves to be an essential ingredient. In addition, we find that the electrons interfere constructively along the two possible paths. In our model, the direct and indirect couplings between the impurities result in an antiferromagnetic spin-spin correlation between them. This interaction, although producing a splitting in dC, as in the experiments, is not enough to take the system out of the Kondo regime for the parameter space studied.

We gratefully acknowledge fruitful discussions with C. A. Büsser, L. O. Manuel, A. E. Trumper, C. J. Gazza, and E. Vernek. G.B.M. acknowledges financial support by NSF under Grants No. DMR-1107994 and DMR-0710529, and E.V.A. acknowledges financial support from the Brazilian

agencies CNPq and FAPERJ (CNE).

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